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**SURVEY OF THE LITERATURE:
CONTROLLED GENERATION OF LIQUID DROPLETS**

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PREFACE

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SURVEY OF THE LITERATURE: CONTROLLED GENERATION OF LIQUID DROPLETS

1. INTRODUCTION

The production of liquid drops of a specific size and quantity per unit time is an ancillary problem in many experimental programs. Forming a multitude of drops from large volumes of a liquid is a widely used process commercially, such as in the spraying of crops with pesticides or fertilizers or spraying paint for surface coating purposes. In most industrial applications, the number of drops generated and their size distribution are not well controlled, but the mass (volume) of drops produced per unit time is controlled to meet the industrial needs.

In laboratory applications, however, the criteria often focus on the concentrations and size distributions produced by drop generators, which are used in aerosol dissemination studies, rather than the amount of material which can be dispersed per unit time. As a consequence, various drop generators have been developed to investigate different aerosol systems, such as spray tanks.

The simulation of the droplet characteristics that are produced from such systems thus involves the formation of a reproducible drop size distribution which reasonably simulates the system's from a liquid that simulates the properties of the liquid used in the system, and that produces drops which can then fall freely before hitting a target material, so that the effect of their impact and interaction with the surface can be accurately tested.

This report reviews and evaluates techniques utilized in generating large drops from visco-elastic liquids, which are also referred to as non-Newtonian liquids. Newtonian liquids are those where the shear force per unit area is proportional to the rate of strain, with the constant of proportionality being the coefficient of viscosity (which is temperature-dependent). Non-Newtonian fluids are those for which the above relation does not hold. These include a variety of liquids, such as thixotropic paints and visco-elastic polymers. Solvents may be thickened by introduction of certain additives and may become either Newtonian or non-Newtonian liquids. Similarly, there are also some liquids which are non-Newtonian even without the addition of thickeners.

The minimum droplet size of interest in this review was a diameter of 0.2 mm (200 micrometers). Difficulties arise in the formation of large drops due to the lack of a strong surface tension force in drops of large radii of curvature. Particularly

in cases with viscous liquids, it becomes very hard to design a system to produce such drops. Additionally, viscoelastic fluids create special problems which must be overcome in order to produce drops of desired sizes on demand.

As a rule of thumb, there is a tradeoff between production of a large number of drops per unit time and production of a well-controlled size distribution. Some techniques, such as the La Mer generator, overcome this tradeoff in special cases (i.e., for small drops of certain compositions which can be produced by evaporation-condensation). Sometimes, hybrid techniques can overcome this limitation; e.g., one technique is used to produce large quantities of a broad size distribution, another technique is used that removes all but the narrow size distribution of interest.

In this review, we examine drop generators which have been developed for various scientific purposes. We have been guided by computerized literature searches, our own reprints of articles on drop generators and drop generation techniques, and by our (DRI) in-house experience in designing and fabricating drop generators for similar purposes. Unfortunately, most drop generation methods are only suitable for producing water drops, or drops of similar low-viscosity liquids. Thus, as the techniques are presented, we note those few which have been tested with highly viscous and/or visco-elastic fluids.

In final analysis, however, most drop generators are unable to produce large, viscous or visco-elastic drops with reasonable size reproducibility. Two generators developed at DRI appear to be suitable, however. The drop ejector is capable of generating drops of viscous, Newtonian fluids over a wide size range, while the microfilm technique is capable of generating visco-elastic fluid drops of masses up to 1 gram.

2. APPROACH

This report surveys a large number of drop generators and focuses on techniques which are proven, or appear to be promising, in the production of large drops, including viscous and visco-elastic liquids, for potential application to military test technology.

The survey of drop generation techniques is presented by grouping similar techniques in terms of the physical principles used in drop generation. One fundamental drop generation technique utilizes capillary flow and drop formation at the orifice, while another technique uses a capillary to form a jet of liquid, coupled with the principle of instability of a liquid jet, which was first rigorously presented by Lord Rayleigh. A number of methods are used to create the jet instabilities. Another drop generation technique utilizes atomization by any of a variety of techniques. Other drop generation principles

include the spinning disk and vaporization followed by condensation. At the Desert Research Institute (DRI), experimentation on methods of producing large drops has been ongoing for over a decade, and continues to develop new techniques.

After presentation of the various drop generation techniques, the report investigates the generation of large drops and drops of viscous liquids, both in terms of special requirements and in terms of the principles which have succeeded, or which appear to be able to succeed, in the production of these types of drops.

3. SURVEY OF DROP GENERATION TECHNIQUES

3.1 Capillary Dropper.

Perhaps the simplest type of drop generator is the capillary tube, through which liquid flows due to the hydrostatic pressure of the liquid reservoir (Figure 1). The capillary tube usually consists either of a stainless steel hypodermic needle or is formed from pulled glass tubing.

The liquid drop is formed as the fluid extrudes through a capillary. As the drop grows, the surface tension acts to keep the drop attached, while gravitational forces act to make the drop pendant and to make it more subject to separation from the capillary. Separation may be spontaneous, or may be induced by any variety of techniques. The largest drop which may be generated in this fashion is dependent on the capillary diameter, the liquid density, and the surface tension. For a given liquid, the maximum drop size is determined by the capillary diameter.

The viscosity of the liquid also plays a role in drop formation, since more viscous fluids require greater pressure gradients to force them through small capillaries. Appendix B discusses some of the physical principles that are involved.

Harkins and Brown (1919) investigated gravitational production of drops from a capillary. They found that water drops of 1 mm diameter required capillary tips of about 0.01 mm, while drops of 0.5 mm diameter required tips of 0.001 mm external radius. Therefore, the capillary method is physically limited to production of droplets large than a few tenths of millimeters diameter. However, smaller drops could be produced by blowing air past the orifice while the water flows through the capillary, producing droplets before they attained the size governed by gravitational separation. In this manner, they produced drops down to 0.1 mm diameter.

Lane (1947) advanced the technique of Harkins and Brown by improvising an apparatus to produce droplets of uniform size. The apparatus (Figure 2) produced drops by blowing air coaxially past a downward pointing hypodermic needle through which the liquid flows under gravity. Careful control of air pressure and liquid pressure gradient through the capillary are required to

produce uniform drops. Lane produced drops between 0.325 and 0.977 mm diameter, with fluctuations in diameters of less than 3 percent.

Reil and Hallett (1969) utilized a device similar to Lane's for producing uniform water droplets of .4 to 2 mm diameter, with a standard deviation of about 3 percent of the mean (Figure 3). Water flowed through a fine capillary and formed at its tip, whereupon a burst of compressed air, directed along the capillary, forcibly removed the drop from the support. The air pulse acted on the drop for less than 0.01 seconds and its pressure varied from .2 to 1 atmosphere, dependent upon the drop size. This technique has the advantage that the drops are removed from the nozzle with a considerable force, but again the factor limiting small drop size is the need to push the fluid through the capillary.

Blanchard (1950) utilized a simple medicine dropper to produce water drops of 5 to 9 mm diameter for study in a vertical wind tunnel. The dropper was calibrated so that drops of known size could be produced with reasonable accuracy. The fluctuations in the masses of drops so produced were probably of the order of 10 percent. Fournier d'Albe and Hidayetulla (1955) used a similar method (described in the section on other techniques) to produce water drops between 9 and 12 mm diameter, for use in studies of drop breakup. Their principal comment on the shortcomings of this technique was the inability to produce uniform size drops, requiring that they compute drop size from the imprints on blotter paper, and the low repetition rate.

The use of a capillary with non-Newtonian fluids involves added difficulties which prevent the reliable production of drops. Non-Newtonian fluids, consisting of long chain polymers or coiled polymers, exhibit extraordinary behavior as a result of their intertwining polymer chains and/or uncoiling and recoiling. As a result, attempts to sever the drop from its support and connecting fluid usually result in the formation of a long, tenuous filament connecting the newly-formed drop and the support.

3.2 Instability of a Liquid Jet.

The previous capillary system is modified into a jet instability system by providing two additional items: (1) a greater pressure to provide a liquid stream, or jet, from the capillary orifice; (2) a device which creates periodic oscillations in the jet.

Lord Rayleigh observed the phenomenon of drops forming from a jet of liquid under the influence of an electric or sonic field in his paper of 1882 and in a companion paper published in the Theory of Sound. He showed that a liquid jet could be stabilized

into drops of equal size and frequency interval by providing some oscillation of the jet, provided that the length of undulation was not more than 4.5 times the diameter of the jet orifice. His theoretical treatment of the forces involved in drop generation has served as the basis for all of the drop generation methods that are based on the breakup of a stream of liquid.

Merrington and Richardson (1947) presented an excellent discussion of the types of breakup a stream of liquid may undergo. The first type of breakup, which they term varicose, is the physical manifestation of Rayleigh theory of breakup of a capillary jet due to the maximum instability of perturbations of length about 4.5 times the diameter of the jet. The second type of breakup occurs when the jet itself is emitted in a sinusoidal pattern, such as through the oscillation of the capillary during emission. This type is called sinuous. This results in the individual portions of the jet being acted upon by air resistance. Attempts have been made to treat this mathematically, but the problem resists analytical solutions. Actually, both the varicose and sinuous types of instabilities occur: at velocities below a critical speed, the breakup is varicose, and at jet speeds in excess of a critical speed, it is sinuous.

A third type of drop breakup, atomization, is covered in section C, below. In this case, a higher critical speed separates this mechanism from sinuous breakup. As Merrington and Richardson explain, it is not only the speeds involved, but other physical parameters which determine how the breakup occurs.

Essentially, breakup by either the varicose or sinuous mechanism is controlled by viscous and inertial forces. Although viscosity does not appear explicitly in the expression describing the varicose type of breakup, it plays a role by determining the rate of fluid response to shear stresses, as Rayleigh (1882) showed.

Experimental studies of varicose breakup by Merrington and Richardson yielded the empirical relation for mean drop size as:

$$V d / [\nu]^{0.2} = 500. \quad (1)$$

In this equation, the mass median drop diameter, d , refers to the size which equally cuts the drop spectrum in two, with respect to mass. V is the relative velocity of the jet with respect to the surrounding air. The kinematic viscosity of the fluid, $[\nu]$, is the ratio of the dynamic viscosity of the fluid to its density. Although this equation is dimensionally inconsistent, it may be applicable to the case of fluid breakup in air (whose kinematic viscosity is constant throughout).

Since the velocity and drop diameter are inversely related, one might expect that larger drops might be generated by providing lower relative velocities. However, this is compromised by a limiting drop size. This critical size was found to be related to the Bond number, which is defined as:

$$Bo = (\rho - \rho') d^2 g / \sigma \quad (2)$$

In this equation, ρ and ρ' are the densities of the liquid and the surrounding medium, g is the acceleration of gravity, and σ is the surface tension of water. The critical size was found to be dependent on Bond number and viscosity, since the viscosity relates the rate at which the fluid distorts in response to external forces. For typical, liquids, the Bond number of the limiting size was found to be about 10. For the viscous liquids investigated, the Bond number of the limiting size was found to be up to 2.5 times greater.

The optimum wavelength for drop formation from a jet differs slightly from the wavelength of maximum instability. Plateau (1874) found that the optimum wavelength for droplet formation was

$$\lambda = 2 \Pi r, \quad (3)$$

where r is the jet radius, λ is the wavelength, and Π is the ratio of the circumference of a circle to its diameter. This investigation is cited by Rayleigh in the study of instability of liquid jets. More recent studies indicate that drops could be easily produced from jets when the wavelength of the imposed oscillation is in the range:

$$7 r < \lambda < 14 r. \quad (4)$$

This range is for wavelengths greater than theory predicts for maximum instability, but perhaps drop formation from the jet is more reproducible when the magnitude of the instability is reduced somewhat. This topic requires additional study for resolution.

Having covered the fundamental theory of drop breakup from a liquid jet, we turn to some of the techniques which are based on this theory.

Dimmock (1950) designed a drop generator from a piece of glass tubing, 3/16 inch in outside diameter, which was heated and drawn

to a tip of about 0.025 inch outside diameter. The capillary reed was vibrated by attaching a steel armature and using a small electromagnet. In this way, continuous streams of drops ranging from 10 to 360 micrometers diameter was produced. For any selected stream of droplets, the size is uniform and the separation between drops is regular.

Magarvey and Taylor (1956) described a mechanical means for generating drops from a stream of liquid. The apparatus, shown in Figure 4, imposes a periodic disturbance on the liquid jet by means of a vibrating earphone. This utilizes the sinuous mechanism of drop formation. The technique was used to produce streams of droplets (about 400 per second) of diameters between 0.3 and 2.5 mm. A second drop technique was used for larger drops (Figure 5). This technique utilizes a vibrating element in contact with the liquid reservoir to impose a periodic pressure pulse on the fluid. By varying the orifice, drops of 2.5 to 10 mm or 10 to 20 mm equivalent diameter could be produced. Their results with large drops, of 2 to 15 mm diameter, indicated that the maximum percentage deviation from the mean diameter was generally less than 0.5 percent; only for the largest drop size was it larger (1.5 percent).

In 1962, Magarvey and his co-workers improved the method of Magarvey and Taylor described above, and added an induction ring beneath the generator in order to impose an electrostatic charge on drops. They were able to produce drops of masses within one percent of the mean, and simultaneously impose identical charges on all the drops in the stream.

Schotland (1960) used a vibrating hypodermic needle to produce drops of 150 to 500 micrometers radius. This technique was improved by Mason et al. (1963), who developed a vibrating capillary device for producing water drops of 30 to 1000 micrometers diameter. The device, shown in Figure 6, utilizes the tuned vibration of the stainless steel hypodermic needle to produce drops. The vibrating diaphragm of the electromagnet is attached to the needle and the apparatus is adjusted by trial and error until a resonance frequency is found, whereupon the tip of the needle vibrates a few millimeters in amplitude. To provide a constant flow through the capillary, an air supply of constant pressure is used. The needle bore diameter determines the drop size range attainable with this technique. For drops less than 160 micrometers diameter, a 5 mm length of 0.1 mm diameter bore was utilized. For drops between 160 and 400 micrometers diameter a 30 gauge needle (bore diameter 140 micrometers) was used. For larger drops, larger needle sizes were used. The drops were produced at a rate of several hundred per second, depending on the resonance frequency of the system.

Sweet (1965) also devised a vibrating needle for droplet production. By charging the drops and subsequently passing them between two plate electrodes, the unwanted drops could be removed from the stream by applying voltage across the electrodes. Eaton and Hoffer (1970) utilized this technique in the production of ethylene glycol drops of 300 to 600 micrometers diameter for the study of terminal velocity and wall effects (Figure 7).

More recently, the oscillations imposed upon the liquid jet have been produced by piezoelectric elements. Fulwyler and Rabbe (1970) imposed fluctuations in the reservoir volume through use of a piezoelectric crystal. Berglund and Liu, in 1973, were also able to use piezoelectric crystals to induce drop formation from the jet. They applied the piezoelectric crystals to the nozzle, resulting in the oscillating orifice. They produced 3 to 20 micrometer diameter drops in this manner.

Thus, a variety of techniques have been used to produce drops through the instability of a fluid jet. Once a stable droplet stream has been established, it is possible to control the number of drops/unit time downstream from the generator. The use of charging and deflecting plates permits the separation of droplets, and hence, provides control of the number of drops that fall in the droplet stream. In this manner, it is possible to charge the drops that are to be deflected out of the drop stream by the deflecting plates leaving the droplets in the stream neutral, or to deflect only the charged drops for use. This procedure allows for experimentation on droplets with known charges.

However, this technique is not very useful with viscous liquids. The principal drawback is the size of the capillary and the force that is required to push the liquid through the capillary. Another difficulty arises with respect to charging the droplet for deflection. Some liquids are not readily charged and will not solvate common salts. Hence, it becomes difficult to separate the drops, making "on demand" generation impractical.

3.3 Atomization.

The techniques devised for the production of aerosols by atomization are so strikingly different than those used for jet instability that their treatment deserves separate sections.

Vonnegut and Neubauer (1952) and Neubauer and Vonnegut (1953) described an apparatus for production of very small droplets from low electrical conductivity liquids by means of electrical atomization (Figure 8). The device uses a capillary, but places

a wire in the reservoir which is connected to high voltage, either AC or positive DC. The droplets produced have diameters 100 micrometer or smaller. In some situations, particles of about 1 micrometer diameter were produced. In general, they found two types of drops produced by electrostatic atomization. The drops formed by smaller electrostatic potentials were larger, and were ejected in uniform streams, thus quite similar to those produced by sinuous disruption of a liquid jet. It is possible that electrostatic energy was utilized in some periodic way to produce this effect. When electrostatic potentials were increased, the investigators noted the presence of "smokes" of water droplets, implying extremely small drop sizes (of the order of 1 micrometer diameter). The characteristics of production of these drops correspond with the atomization of a liquid jet.

Drozin (1955) found that the electrostatic potential applied to the atomizer governs the type of drop. Similar studies were conducted by Zeleny (1914) and Bollini et al. (1975). Nawab and Mason (1958) verified the findings of Neubauer and Vonnegut (1953) that the liquid conductivity plays a role in the ability to form drops by electrostatic atomization.

Ryley and Wood (1963) discuss a vibrating capillary atomizer construction and theory of operation (Figure 9). The Rayleigh criterion ($\lambda/d = 4.5$) is demonstrated, as are the second and third harmonics, which are not revealed in the Rayleigh instability criterion. Additionally, they point out that one cannot arbitrarily select an oscillation frequency, since only harmonics of the fundamental instability frequency produce single drops reliably. Finally, they stress the agreement of experiment with Rayleigh theory regarding the liquid's physical properties (surface tension and density). They note that a generator which works for one liquid will not work, without some setting change, for another liquid of different physical properties. Water drops produced with their vibrating hypodermic needle apparatus were controllable in the range 300 to 1350 micrometer diameter.

Hoffer and Perthel (1968) developed an aerosol generator which sprays liquid solutions as droplets of 5 to 10 micrometers diameter, from which the desired aerosol is obtained by solution evaporation. The drop size distribution was found to be quite stable, resulting in the generation of a monodisperse aerosol.

Brownscombe and Hallett (1967) produced a narrow size distribution of water drops using the vibrating needle technique of Mason et al. (1963). They produced drops of 9, 19 and 40 micrometers radius with standard deviations of 1, 2, and 7 micrometers, respectively by vibrating a metal rod longitudinally at frequencies of 100, 40, and 13.5 kHz, respectively. The liquid flow rate was less than 0.25 milliliters per minute.

Subsequent experiments, following the method of Brownscombe and Hallett (1967), used a rod vibrating at 100 kHz to produce drops in the size range of 5-10 micrometers. The drops could be generated with a stable distribution, provided that the liquid flow was extremely well controlled and that the liquid was always introduced onto the vibrating rod at the same point.

In summary, drop generation by atomization is not well-suited for viscous liquids. The liquid is forced through a fine capillary using a constant flow pump that exerts minimal force. Even if the liquid flow problem could be alleviated, the vibrational energy in the needle or steel rod would be insufficient to breakup the non-Newtonian fluid.

3.4 Spinning Disk.

The use of spinning disks for droplet generation has been described by Walton and Prewett (1949) (Figure 10) and modified by May (1949). Essentially, a stream of water on the upper surface of a smooth, completely wetted disk spinning with its perimeter traveling at about 100 mph produces drops by centrifugal action. Although the spinning disk method produces a broad size distribution, the drops fall in different trajectories due to their different sizes. Thus, for any specific trajectory, the drop size distribution is approximately a normal distribution with a narrow standard deviation. Drops of 10 to 250 micrometers diameter have been produced in this fashion. The spinning disk is actually a spinning cone, utilizing a technique described by Beams (1937) for rapidly spinning a cone using the Bernoulli principle.

Walton and Prewitt (1949), Beams (1937), and others have described the theory and experimental apparatus that permits the generation of many drops of a uniform size using the principle of centrifugal force. In this instance they have found for water that:

$$d = 2.78 (\sigma/D)^{0.5} / \Omega \quad (5)$$

where d is the drop diameter (cm); σ is the surface tension of the liquid interface (dyne/cm); D is the disk diameter; and Ω is the angular velocity of the disc.

The centrifugal force on the drop at radius R from the axis of rotation of the disk is

$$F = m v^2 / R = m R [\Omega]^2 \quad (6)$$

This is counteracted by the attractive surface tension force

$$F = k \sigma D \quad (7)$$

This yields the drop diameter D

$$D = k \sqrt{\sigma / (2 R \rho)} / \Omega. \quad (8)$$

Walton and Prewett (1949) found that

$$2.67 < \Omega D \sqrt{2 R \rho / \sigma} < 4.44 \quad (9)$$

and Phillipson found that

$$2.64 < \Omega D \sqrt{2 R \rho / \sigma} < 3.06 \quad (10)$$

In Walton and Prewitt's apparatus liquid was introduced through a nozzle onto the center of a spinning disc. The liquid flowed outward over the wettable disc until it reached the outer edge. There it was formed into drops whose mean size is given by the above equation. In use, the drop generator had a housing over the disk to limit the angle drops exit the generator. Drops are produced over 360 degrees but the housing defines the area through which the drops can be ejected. All of the drops are not the same size. However they can be readily sorted into uniform sizes by distance from the generator. Larger drops will fall onto a surface closer to the generator than smaller ones. Since the drop size is related the angular velocity, theoretically any size drop can be produced. In practice, a spinning disc driven by a variable speed motor has been used for to generate drops as small as 70 micrometers in diameter. In Brown's apparatus using a spinning bucket, 70 micrometer diameter drops were produced at 10,000 rpm. Smaller drops can be produced at higher speeds by driving the disk with compressed air. In this case however the entire apparatus must be enclosed in a sturdy housing due to the possible fragmentation of the disk.

Although this type of generator has some faults, for example, multiple droplets at uncontrolled positions and the potential for contamination of the liquid by the metallic disc, it does produce drops over the entire size range with minimal restrictions on the type of liquid. Viscous liquids however will tend to wet the surface of the disk, making generation difficult. In Brown's apparatus the non-Newtonian liquid will spin off a web of long, slender threads.

A technique similar to the spinning disk generator has been developed. This uses a rapidly spinning bowl containing the liquid, with a small bore hole drilled near the bottom for the liquid to escape (Figure 11). As the bowl is rotated at 15,000 rpm, the liquid stream produces droplets as it emerges from the bowl. The drops produced have diameters ranging from 80 to 400 micrometers, depending on the bore size of the hole and the angular velocity of the bowl.

Watson (1948) showed the effects of pressure and chamber dimensions of a swirl atomizer on the drop size. Generally, smaller chambers produced smaller drop sizes. In general, however, dribbling prevents generation of a narrow drop size distribution.

3.5 Vaporization-Condensation.

Sinclair and La Mer (1949) devised an improvement of the La Mer aerosol generator (Figure 12) for the generation of uniform liquid droplets. A sodium chloride condensation aerosol is produced and mixed with the boiling solution. The airstream is directed to a reheater unit, and the growth of the drop proceeds by rapid condensation as the airstream exits the unit through an inverted outlet tube and is rapidly cooled (Figure 13). Monodisperse drops of up to 4 micrometers diameter were produced in this fashion. Drops of 2 to 58 micrometers radius were produced by Burgoyne and Cohen (1953) by making additional improvements to the inverted outlet version of the La Mer generator.

Thickeners, especially non-Newtonian polymers, have properties that will make drop generation using these techniques improbable. Unless the thickener condenses at the same temperature as the solvent, it is not possible to grow the drops by condensation. Hence, these techniques were not considered further in specifying drop generators for viscous liquids.

However, for future consideration, it is useful to envision a two-stage La Mer generator. The first stage might condense the higher boiling-point substance, probably the thickener, to form a monodisperse aerosol of one substance, which is introduced into a controlled-temperature chamber with saturated solvent vapor, causing the aerosol particles to grow by condensation until they consist of viscous solution drops. This concept could be adapted to non-Newtonian solutions, provided of course that the thickener could be condensed onto an aerosol nucleant, and subsequently serve as a nucleant for the solvent. Additionally, the question of whether such a process could dissolve the thickener in the solvent would need to be investigated.

3.6 Impulse Generation.

Fournier d'Albe and Hidayetulla (1955), in their study of the break-up of large water drops, produced drops by drawing a known quantity of water into a 6 mm internal diameter glass tube fitted with a rubber bulb on one end, and then applying sudden pressure on the bulb. They report that this technique occasionally produced one or more satellite drops. The drops thus produced, ranging from 8.5 to 12.5 mm diameter, were sized by adding the masses of the break-up drops produced in the experiment.

Although these devices use capillaries that are large with respect to those more commonly used by other investigators, it does not appear that highly viscous liquids can be readily forced through the system. The impulses that are applied are not of sufficient magnitude to form drops from a liquid jet.

Hoffer and Pitter (1982) fabricated a generator for large drops that hydraulically ejects the fluid from a tube. The apparatus, as slightly modified in 1987, is shown in Figure 14. A syringe plunger is filled with iron filings and acts as the armature of a solenoid. The principal advantages of this generator are the absence of capillaries, the ability of the generator to produce a single droplet which may be positioned in the x-y plane, and the large amount of impulse which can be applied to eject the liquid from the reservoir. However, even by applying a large impulse, the drop generator cannot produce drops of non-newtonian liquids. Figure 15 details the nozzle design for a two-fluid generator, using a hydraulic fluid in most of the lines, and the thickened fluid only at the nozzle. Drops of non-Newtonian liquids exit from the generator, but do not cleanly separate, and hence they spin long strings as they fall. The characteristics of this generator are thought to be those that must be used if the problem of generating large, viscous drops on demand is to be solved. These are: design to produce single drops; ability to apply a large impulse consistently, drop after drop; and no capillaries. In the subsequent discussion of the improved drop generator these are discussed further even though a final solution has not been reached.

3.7 Other Techniques.

Rayner and Hurtig (1954) utilized a novel variation on the theme of separating drops from a fluid stream for toxicological studies. A capillary needle provides the liquid feed, and a vibrating coping saw blade, which has been shaped so its end is pointed and sharpened at the edges, is placed immediately under the needle orifice, so that the blade's oscillations withdraw filaments of the liquid from the emerging drop at the orifice (Figure 16). The filaments break off of the blade, forming two streams of uniform-size droplets of 70 to 400 micrometer in diameter. Difficulties with this technique include the dependence on liquid feed rate and the velocity and frequency of vibration of the blade tip as it passes through the feed drop.

Rayner and Haliburton (1955) improved the vibrating blade technique by designing a rotary blade drop generator (Figure 17). A spindle is driven at 600, 900, or 1200 rpm and stabilized to a constant rotation rate. A flat steel blade is attached to the spindle and adjusted to cut through the feed drop. This apparatus produces uniform drops whose size (mass) are linearly related to feed rate (hydrostatic head height) for a given liquid and blade rotation rate. Certain liquids have a tendency to stick to the blade surface, creating more dispersion in the size distribution. This problem is eliminated by suitably coating the surface of the steel blade.

However, mechanical slicing of a viscous drop is not an acceptable technique unless one can ensure that the liquid will not adhere to the blade. If any liquid adheres to the blade, this will result in production of threads when using visco-elastic fluids.

Liu (1967) investigated a method of drop generation based on the principle of aerosol cans, that of liquified gas atomization. The study, however, found that the drop size distribution was not controllable by can pressure, and although most drops emitted were in the sub-micrometer size range, the generator produced a broad size distribution.

Another interesting device is the whistle atomizer. This is a sonic atomizer system, so named because of the 10 kHz oscillation imposed to break up the drops. It has been found that the drop size distribution is mostly controlled by the physical dimensions of the atomizer, and fine control cannot be achieved through variation in liquid or air feed rates or pressures.

Blanchard (1954) demonstrated a method for producing very small water drops by the bubble burst mechanism (Figure 18). Although the sizes of drops produced in this fashion are variable, the height to which any size drop reaches as a result of the upward moving jet is remarkably constant.

Binek and Dohnolova (1967) investigated the production of drops by periodically dipping a whisker into a reservoir of fluid and withdrawing it. Surface tension would adhere the liquid to the whisker, and upon withdrawal it would extract a filament of liquid from the reservoir. The filament, initially approximating a long circular cylinder, would breakup and form either a single drop or a larger drop with one or more smaller satellite droplets.

Abbott and Cannon (1972) devised an improvement in a water droplet generator (Figure 19). They followed the fundamental design of Wolf (1961), who constructed a metal wire attached to a vibrating reed, but where Wolf had used a tuned vibrating reed to

continuously produce drops of one drop size, Abbott and Cannon implemented electronics to control the arm movement and drop charging.

The National Aeronautic Establishment, of the National Research Council of Canada recently devised a drop generator for producing drops of 30 to 800 micrometers diameter from water or heavy fuel oils. The standard deviation of drop size was less than one percent of the mean. The device is similar to that of Abbott and Cannon (1972), using a thin needle glued to a speaker to oscillate into and away from a pendant drop hanging from a vertical capillary (Figure 20). The filament of liquid thus extracted from the drop would breakup into a small, freely falling drop. Unfortunately, studies of this device with viscoelastic fluids indicated that it could not operate with such liquids.

Sayler (1985) devised a novel hybrid drop generator, called the DICE (Drop Injection Clean Entry), which atomizes a viscous liquid by use of another fluid (liquid or air) which is immiscible with the first. Drop separation is accomplished by separating the drop volume from the rest of the fluid in the capillary by means of an internal bubble of the second fluid. This is provided by a small diameter hypodermic needle inserted through the capillary wall near the exit (Figure 21). Using this device, non-Newtonian liquid drops (diethyl malonate, thickened with 5 percent by weight K-125) of 2 or 3 millimeters diameter were produced using air bubbles. The minimum size which could be produced was about one millimeter diameter, since smaller drops would not cleanly separate from the capillary. Somewhat larger drops than were investigated could conceivably be produced. When an immiscible liquid was used for separation, a satellite drop of the separation liquid was often co-generated. For testing purposes, a means for removing the separation fluid droplets is required in order to prevent their interference with the collection of or behavior of the primary droplets.

The Desert Research Institute has recently developed a novel technique for generating large, viscous and viscoelastic fluid drops. The technique is illustrated in Figure 22. It involves suspension of the drop on a microfilm attached to a ring. The microfilm, which is soluble in acetone, is immediately disintegrated when sprayed with a small amount of acetone, causing the drop to fall. The residual microfilm on the surface of the drop is but a small fraction of the drop volume, and the acetone rapidly evaporates, so the effect of the suspension and release agents is minimal on the interaction physics which occur when the drop strikes a surface.

4. DISCUSSION OF GENERATION OF LARGE DROPS

Drops of diameters greater or equal to 0.2 mm (masses in excess of about 5 micrograms) can be generated by a variety of techniques. However, most techniques have been applied to water or aqueous solutions.

Large drops are not easily created with a great degree of uniformity. The large mass of such a drop cannot be supported by surface tension on a capillary tube, and thus requires more elaborate formation techniques. With regard to the Rayleigh instability technique for generating drops, the large radius of curvature negates any role of surface tension towards stabilizing perturbations and controlling drop size. Atomization techniques produce drops of less than 100 micrometers diameter.

The evaporation-condensation technique employed by La Mer and others utilizes the principles of condensation growth, wherein the dimension of the drop increases as time to the $2/3$ power and as supersaturation vapor pressure. Difficulties in maintaining a large supersaturation and a sufficient volume for drops to grow (as they fall) for sufficiently long times prohibit the formation of suitably large drops.

The task of reproducibly creating large drops is thus one of devising some physical means by which a large proto-drop first can be suspended until it is ready for dropping, and second can be severed cleanly from the support without formation of satellite droplets. Drop mass is generally of primary concern, since it provides the hydrostatic force which often limits maximum drop size. Surface tension is also of concern, since it provides the suspension force in capillary systems.

Capillary droppers are capable of producing water drops up to a few millimeters diameter, and monodisperse drops of 0.3 to 1.0 micrometer diameter can be produced by introducing a coaxial air puff (Lane, 1947). Both of these methods can generate either single or multiple drops, and the latter technique has good control of drop size.

Medicine dropper-like glass tubes with rubber bulbs have been used to generate extremely large, single water drops by Blanchard (1950) and by d'Albe and Hidayetulla (1955). These drops, which can exceed a centimeter in equivalent spherical diameter, have rather poor size control (errors of about 10 percent) in contrast to other techniques with size control of about 1 - 3 percent.

Techniques which utilize Rayleigh instability of a liquid jet produce a variety of drop size ranges. Dimmock (1950) produced water drops up to 350 micrometers diameter, while Magarvey and Taylor (1956) were able to produce water drops up to 2.5 millimeters and 2 centimeters diameter by imposing oscillations on the liquid reservoir. These techniques produce more or less steady streams of drops.

Various researchers have imposed resonant vibrations on hypodermic needles to produce streams of water or aqueous solution drops, perhaps electrically deflected to or from a target (Schotland, 1960; Mason et al., 1963).

It can be stated that, generally, these drop sizes are well-controlled, and may be selected from a few micrometers to over one millimeter diameter. In one example of a distinctly different solution used in such a device, Eaton and Hoffer (1970) produced ethylene glycol drops up to 600 micrometers in diameter using this technique.

Spinning disks have been successfully applied to produce drops up to about 250 micrometers diameter (Walton and Prewett, 1949; May, 1949). The limitation of spinning disks lies principally in the role of viscosity in droplet formation. Thus, the technique is not suited for high viscosity liquids.

Reil and Hallett (1969) utilized a strong pulse of compressed air to blow a drop off a capillary. They produced drops of 0.4 to 2.0 millimeters diameter with good reproducibility.

The DICE, developed by CRDEC, provides excellent drop size control from about 1 millimeter diameter to 3 and possibly 4 or 5 millimeters diameter. It uses a capillary system to suspend a pendant drop, then it forms an air bubble near the exit to cleanly sever the drop from the remainder of the fluid. This is highly reproducible and has been found to work with a thickened, non-Newtonian fluid, DEM (diethyl malonate) thickened with 5 percent by weight of K-125.

The large drop generator developed by Hoffer and Pitter (1984) and more recently modified to permit greater reproducibility and drop size consistency utilizes a hypodermic syringe positioned in a solenoid, with its plunger filled with iron filings acting directly as the armature, to provide a rapid, controlled volumetric pulse to the closed liquid reservoir, ejecting the drop on a parabolic trajectory upwards. Drops produced in this manner had diameters which were reproducible within 1 to 3 percent in the range of 0.3 to 1.2 centimeters diameter. More importantly, as discussed in more detail in the following section, the drop generator is designed for producing large drops from viscous liquids. In recent experiments, it was determined that certain viscoelastic fluids could not be used, because they did not separate to form discrete drops on ejection.

The other DRI drop generator of interest was recently devised to generate large, viscoelastic liquid drops. It consists of a microfilm mounted on a frame onto which a liquid drop is extruded. The microfilm can support drops in excess of 0.5 grams mass. When sprayed with acetone, the microfilm disintegrates and the drop begins its fall. A small amount of microfilm remains attached to the drop surface as a small, insoluble lump, but it probably plays a minimal role in drop aerodynamics or splashing/penetration physics, because of its small mass relative to that of the drop, and because it moves to the top of the falling drop as a result of its lower density. Any acetone which comes into contact with the drop rapidly evaporates, and does not affect the drop's fall or splashing characteristics.

5. DISCUSSION OF GENERATION OF VISCOUS AND VISCO-ELASTIC DROPS

Only a few of the generation techniques described in this report have been tested with non-Newtonian liquids. However, most utilize physical principles which greatly restrict the use of either viscous or visco-elastic fluids. Viscous fluids are not well suited for capillary systems; their viscosity results in a large resistance per unit length of the capillary system, requiring high pressure to force the fluid through. Viscous fluids are also not well suited for other types of generators, such as atomizers, sprayers, or spinning disks.

The Rayleigh instability criterion has not been carefully investigated with thickened liquids. Since it is based on inviscid liquid breakup, there is probably some correction which would apply to the familiar breakup criterion; there is evidence of this correction in studies of not-so-viscous fluids.

Visco-elastic fluids require even more consideration for development of drop generators. By their nature, they cannot be separated by applying high shearing stresses; they require a slow, steady application of shearing stress to successfully separate. This time constraint becomes more limiting with larger drop sizes, because of the difficulty in keeping larger drops suspended for longer times.

For viscous fluids, the DRI drop generator (Hoffer and Pitter, 1984) produces large drops with good reproducibility. It also produces a range of drop sizes from about 1 millimeter diameter to about 12 millimeters equivalent spherical diameter. However, the ejection technique does not work with highly visco-elastic liquids.

The DICE (Sayler, 1985) is an excellent technique for producing visco-elastic drops of 1 to 4 or 5 millimeters diameter. It overcomes the visco-elastic nature of the fluid by relatively slow expansion of an air bubble in the capillary near the end, separating the drop from the reservoir fluid. This technique is greatly superior to air blasts applied externally to the capillary.

For larger drops, the only technique so far demonstrated useful for visco-elastic drops is the microfilm technique currently under development at DRI. This technique is capable of supporting liquid drops of a gram or so, and releasing them into vertical fall by application of a spray of acetone, which rapidly dissolves the membrane.

While this survey has identified only three design options for generating large viscous or visco-elastic droplets, these designs are applicable to the range of size and viscosity which is of interest to aerosol test technologists.

6. SUMMARY AND CONCLUSIONS

The majority of drop generators discussed in the literature are designed for production of water drops, and make use of the low viscosity (low-resistance capillary flow), high interfacial surface tension with glass (adhesion to capillary during drop formation), and high surface tension (reproducible formation of uniform size drops at diameters less than a few millimeters). The techniques in use, such as capillary flow, oscillation of a jet, and filament extraction with a fine wire, are not well suited for forming drops from highly viscous, or visco-elastic liquids.

The survey identified only three techniques which have demonstrated the capability of generating large, viscous or visco-elastic drops:

A.) The Desert Research Institute's drop generator which is based on drop ejection in response to an impulse imposed on the fluid reservoir; it does not perform very well with visco-elastic liquids.

B.) The CRDEC DICE drop generator for visco-elastic liquids, in which an immiscible fluid is injected to force separation of the primary fluid from a pendant drop. The generator is effective for drops up to about 4 or 5 millimeters diameter.

C.) The Desert Research Institute's microfilm technique for use with drops exceeding about 0.5 grams mass. The reproducibility of this technique is dependent upon the method of extrusion of viscous drops, not on the release method.

These techniques presently exist as one-of-a-kind research devices. Further studies would be required to fully demonstrate their capabilities and to adapt them to routine use.

The unifying concept of these three drop generators is their avoidance of the drop formation methods most commonly employed by water drop generators. Based on this observation it is concluded that additional, effective generators for large, viscous drops can yet be invented. The key to their successful development is the application of novel techniques in response to the physical characteristics of the liquid of interest.

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APPENDIX

CAPILLARY DROP PHYSICS

For any given liquid, there exists a maximum drop size which can be supported. This is because the capillary diameter not only controls the surface tension force, but also controls the rate of fluid feed to the drop.

The use of capillaries for thickened liquid drop generators is limited. Although the viscosity does not enter into the equations for gravitational or surface tension force, it enters into the overall system by means of the reduction of pressure through the capillary. For Hagen-Poiseuille flow through a circular cross section pipe, the shearing stress is inversely proportional to the Reynolds number, and the pressure gradient is given by

$$dp/dz = 64 R \rho v_{av}^2 / (Re) \quad (A-1)$$

When the Reynolds number (Re) is expanded, the equation becomes

$$dp/dz = 32 \rho \nu v_{av} \quad (A-2)$$

or, using $\nu = \xi / \rho_f$,

$$dp/dz = 32 \rho \xi v_{av} / \rho_f \quad (A-3)$$

An assessment of maximum drop size may be derived from consideration of the force balance at the capillary/drop interface. The interface has length of $2 \pi R_c$. As a consequence of the geometry of a drop of radius R_d , the angle θ between the line of contact and the horizontal is:

$$\theta = \arcsin (R_c / R_d) \quad (A-4)$$

The upward force holding the drop to the capillary is then:

$$F_u = 2 \pi R_c \sigma_{lv} \sin \theta \quad (A-5)$$

The downward force of the drop is the weight of the drop below the capillary orifice:

$$F_d = \pi \rho R_d^3 (2/3 + \cos \theta - \cos^3 \theta / 3) \quad (A-6)$$

As long as the upward force exceeds or equals the downward force, the drop remains attached to the capillary. A simplified form of the balance states that the maximum suspended drop radius which can be supported is

$$R_d^4 = 3 R_c^2 \sigma_{lv} / (\alpha \rho) \quad (A-7)$$

where α is somewhere between 1 and 2. The mass of the detached drop is dependent on α , since the truncated sphere is assumed to be detached:

$$\text{Mass} = 2 \alpha \pi \rho R_d^3 / 3. \quad (A-8)$$

Thus, assuming α is close to 2, the largest drop radius is proportional to the square root of the capillary radius and the fourth root of the interfacial tension. Thus, for practical applications, the capillary size determines the range of drop sizes. Even if a drop is not too large to be supported, it may be generated by application of some force (a burst of air or vibration of the capillary).

However, as the capillary becomes very large, it becomes more and more difficult to control the fluid extrusion rate, and thence to control drop sizes. In the other limit, as smaller capillary sizes are used, the maximum drop size can be more easily limited, but the force required to extrude the liquid at a reasonable rate increases dramatically.

Consideration of minimum drop size may be derived from examination of pressure balance in the drop. Assuming a spherical drop of radius R_d suspended from a capillary, the pressure difference across the drop interface is:

$$p_d - p_a = 2 \sigma_{la} / R_d \quad (A-9)$$

Similarly, the pressure difference between the "head" and the drop is:

$$p_d - p_h = \rho g h + (dp/dz) dz \quad (A-10)$$

where h is the height of the head from the drop and dz is the capillary length. Equations 9 and 10 may be combined to solve for the drop size:

$$R_d = 2 \sigma_{la} / (p_h - p_a) + \rho g h + (dp/dz) dz). \quad (A-11)$$

In the denominator on the right hand side, the minimum value which can be attained occurs when (dp/dz) goes to zero (negligible velocity or very slow flow) and $p_h + \rho g h$ is close to, but slightly greater than, p_a . (The terms must be greater than atmospheric pressure or the drop is sucked back into the capillary.)

The pressure gradient is independent of the capillary radius, R , but is proportional to the fluid viscosity and the average velocity of the fluid through the capillary. Fluids with high viscosities require large pressure gradients to force the fluid through the capillary. This may be addressed with short capillary lengths and/or slow extrusion rates.

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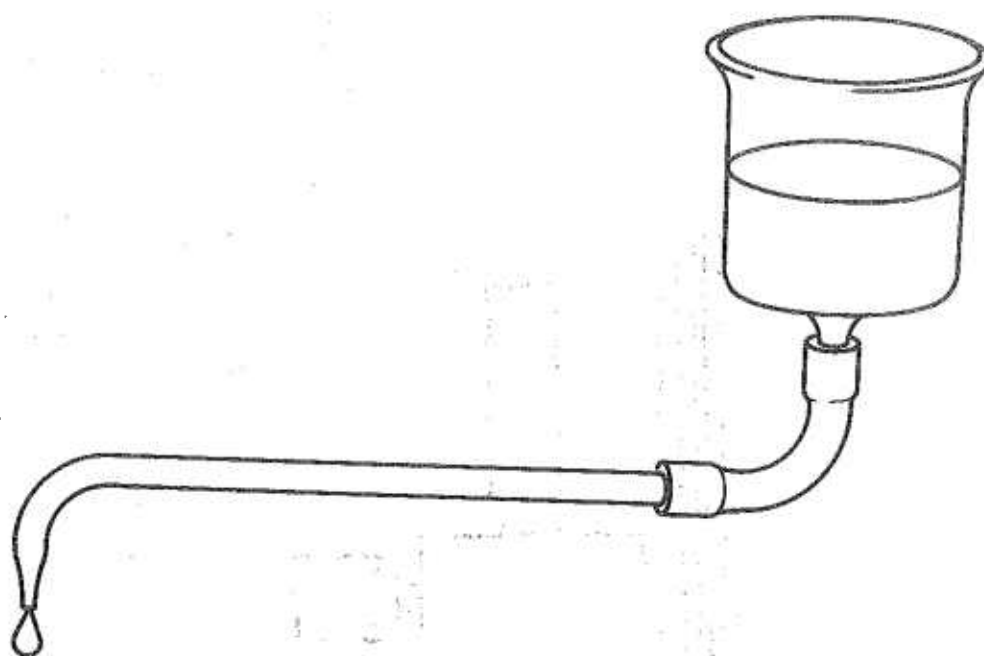


Figure 1. A Simple Capillary Tube Drop Generator.

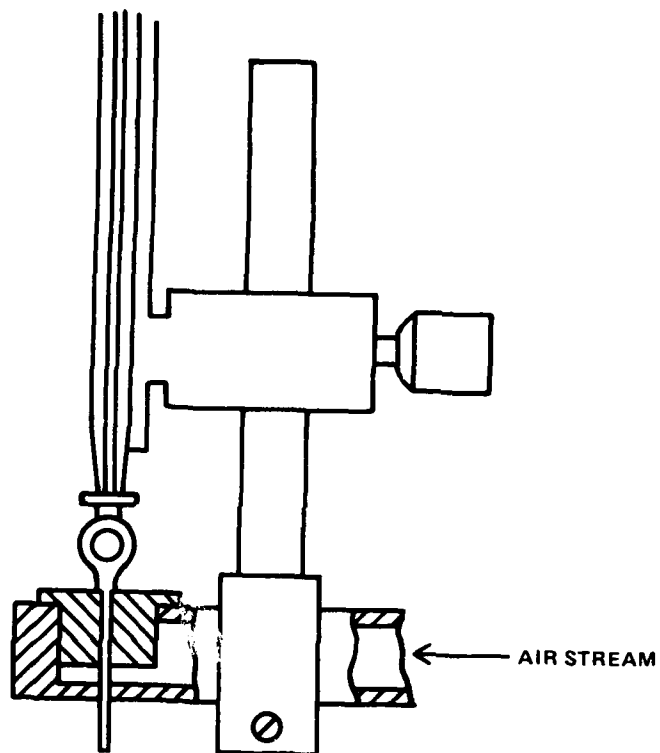


Figure 2. Apparatus of Lane (1947) for Producing Uniform Drops, Using an Airstream to Separate Drops from the Capillary.

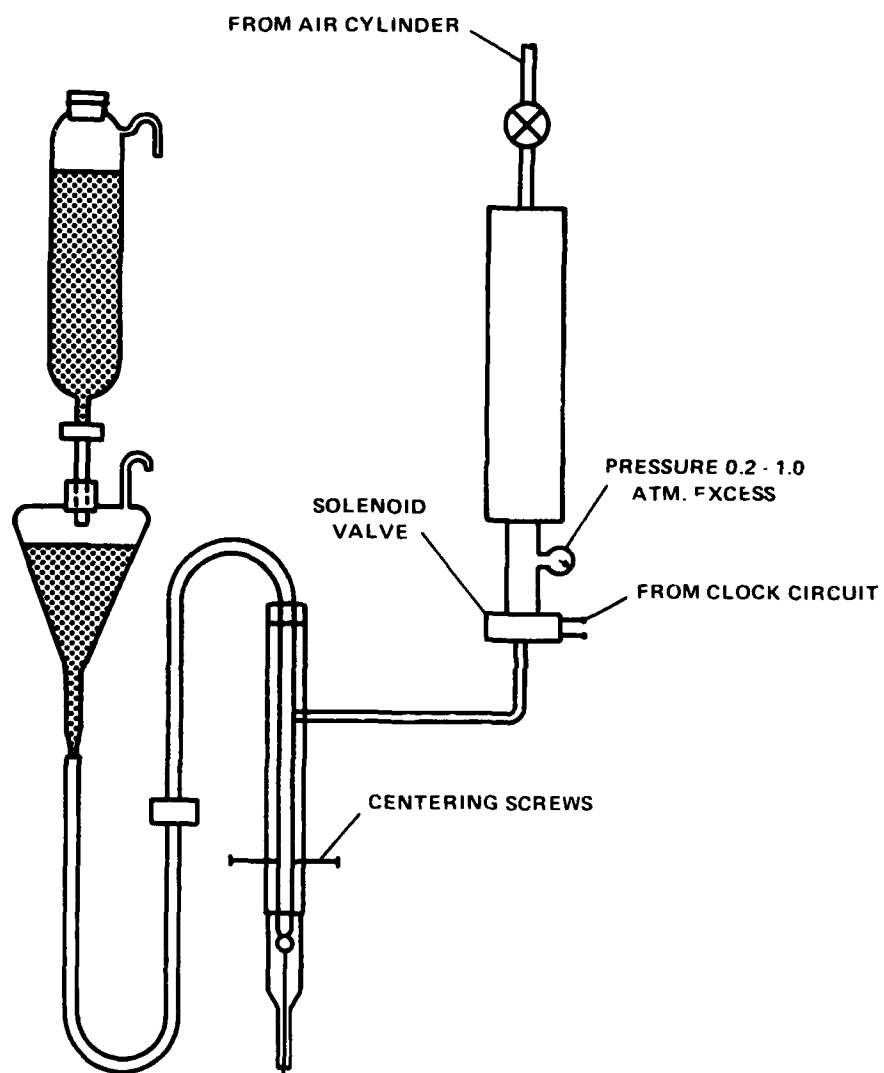


Figure 3. Apparatus of Reil and Hallett (1969) for Production of Uniform Drops Using Timed Pulses of Air.

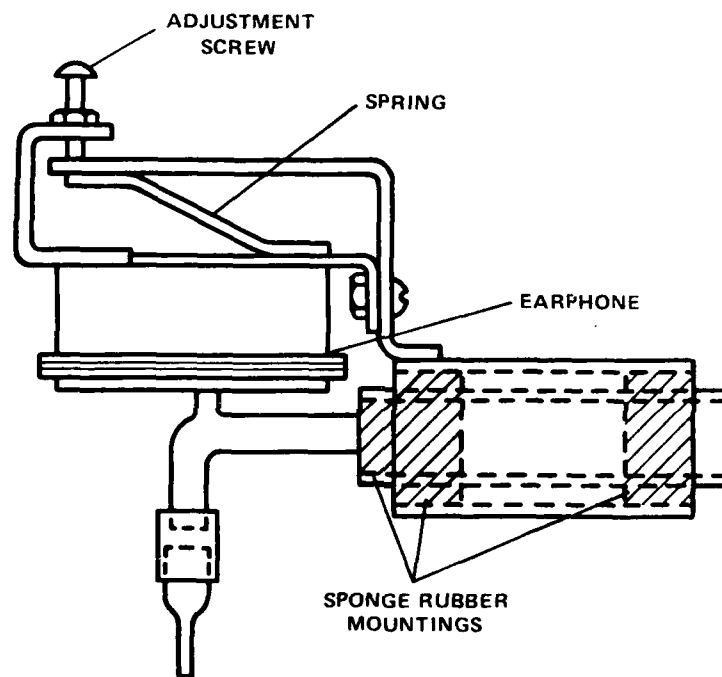


Figure 4. Apparatus of Magarvey and Taylor (1956) Showing Technique of Imposing Vibrations on the Capillary.

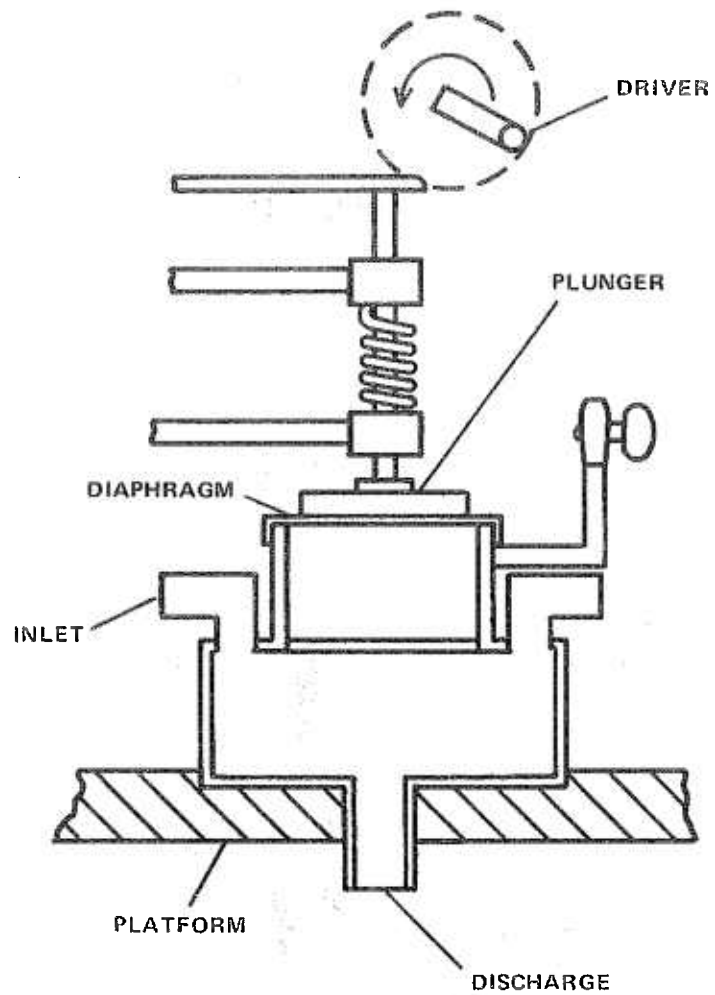


Figure 5. Technique of Magarvey and Taylor (1956) for Imposing Pressure Oscillations on the Fluid Reservoir.

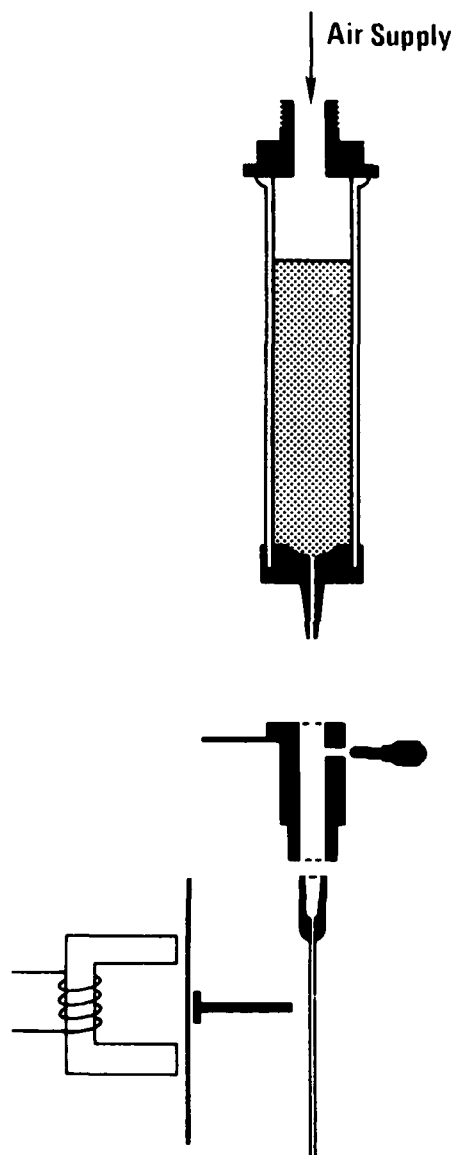


Figure 6. Apparatus of Mason et al. (1963) Using a Vibrating Stainless Steel Hypodermic Needle.

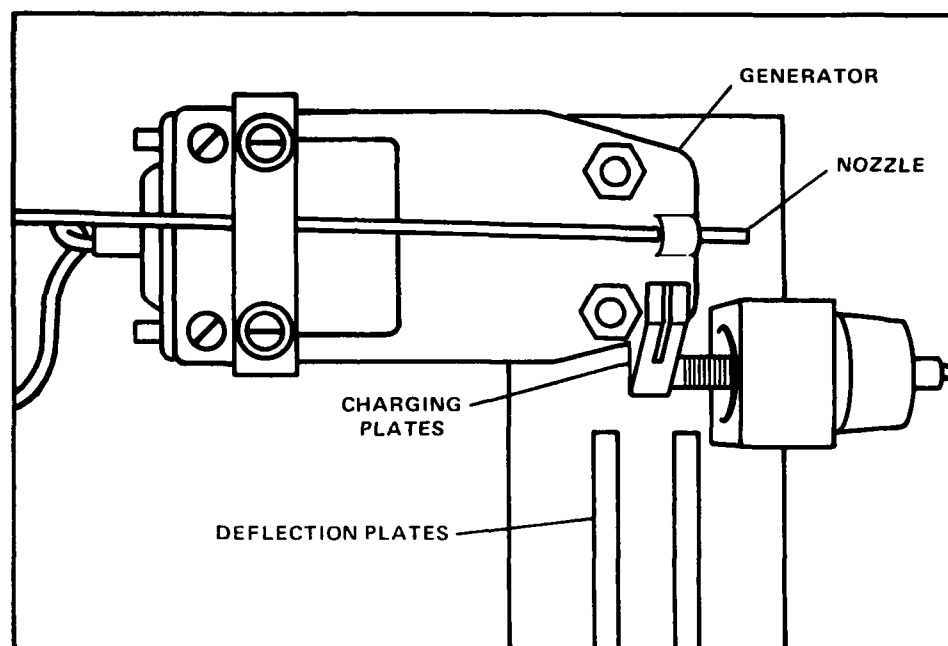


Figure 7. Apparatus of Eaton and Hoffer (1970) Using a Vibrating Hypodermic Needle and Drop Charging Plates.

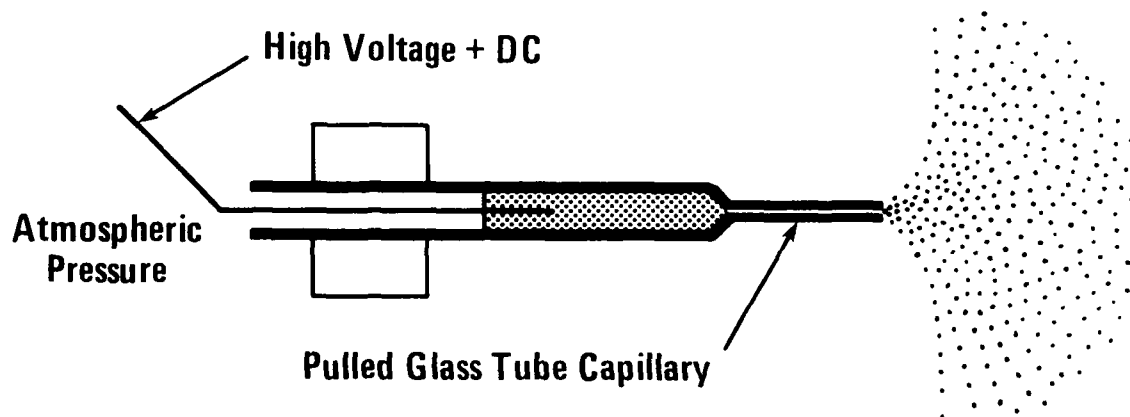


Figure 8. Apparatus of Neubauer and Vonnegut (1953) for Electrostatic Atomization.

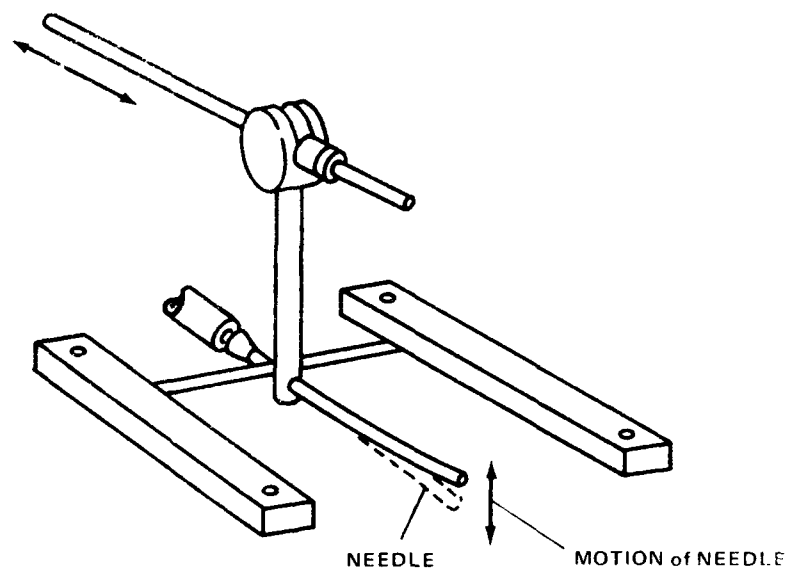


Figure 9. Apparatus of Ryley and Wood (1963) for Mechanical Atomization.

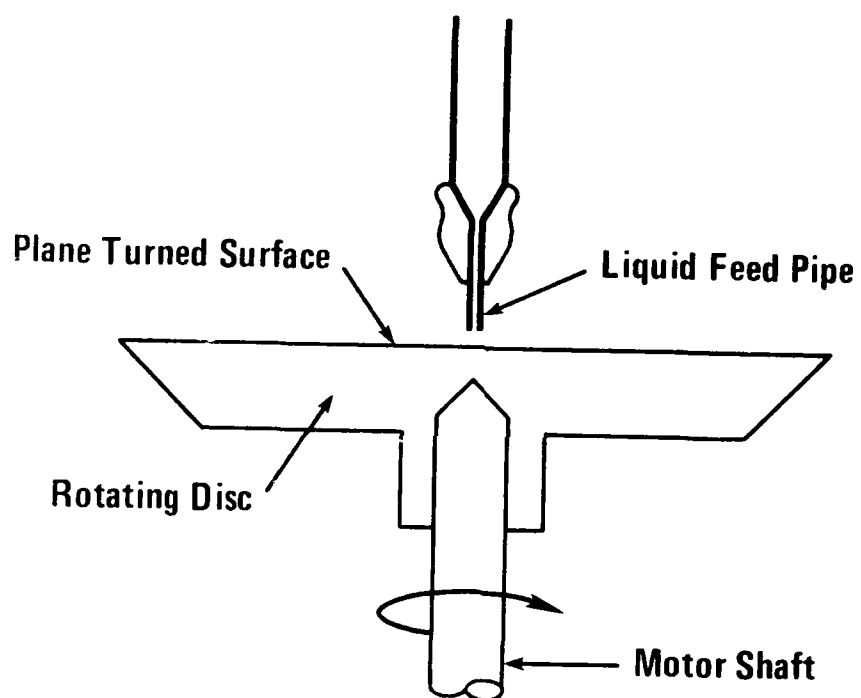


Figure 10. Design of Walton and Prewett (1949) for Spinning Disk Drop Generators.

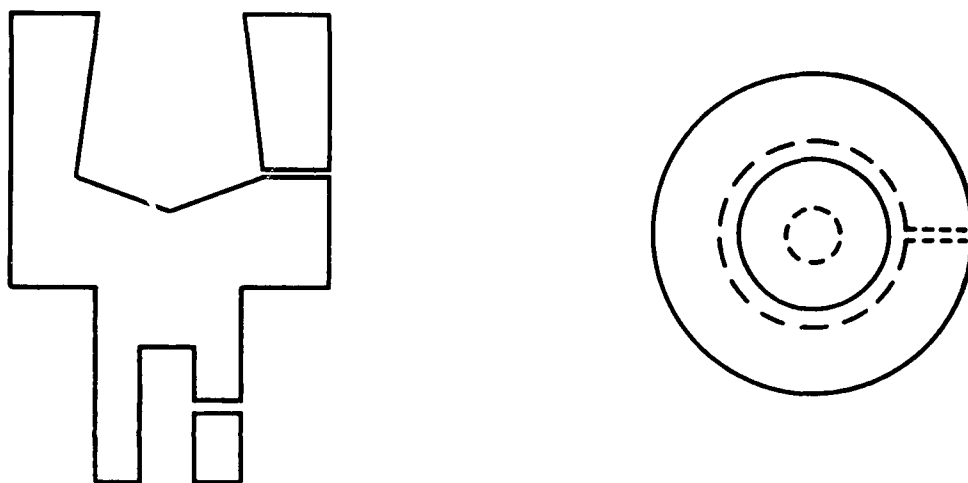


Figure 11. Apparatus for the Production of Drops With a Rotating Bowl.

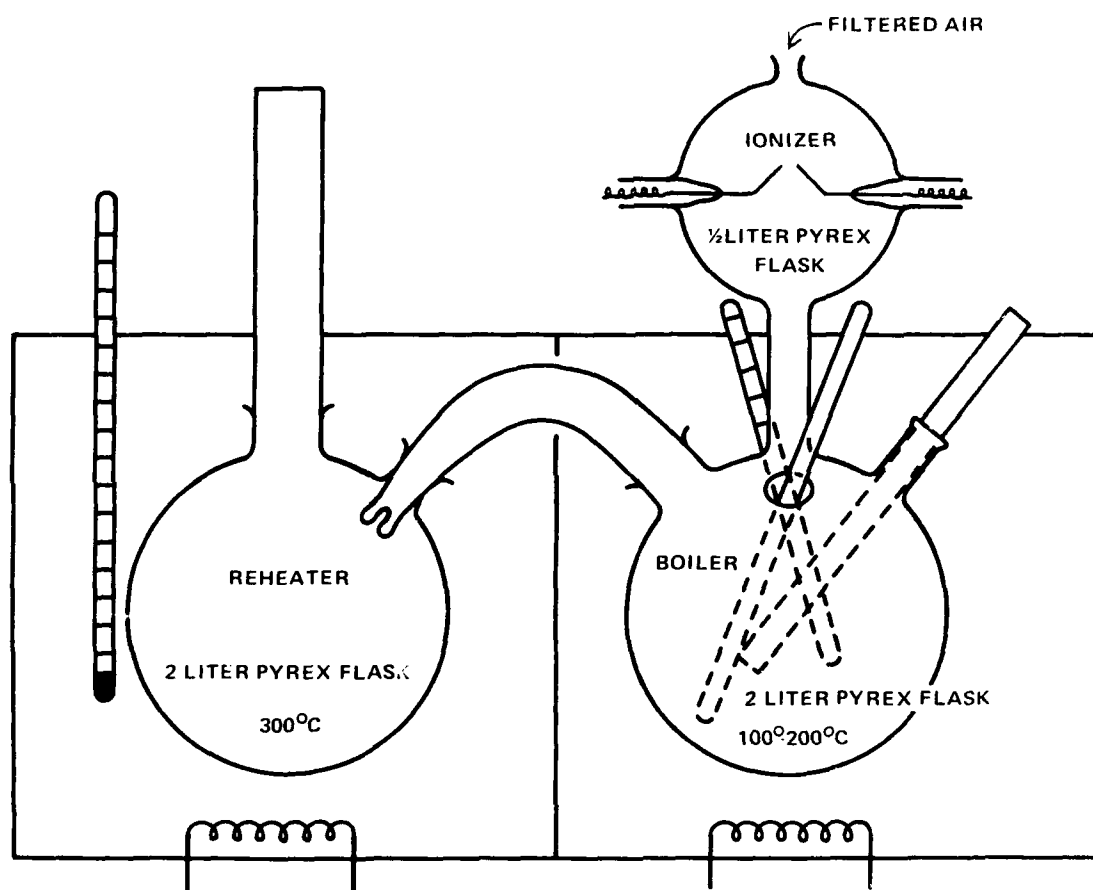


Figure 12. La Mer Generator for Uniform Liquid Drops and Aerosol Particles, from Sinclair and La Mer (1949).

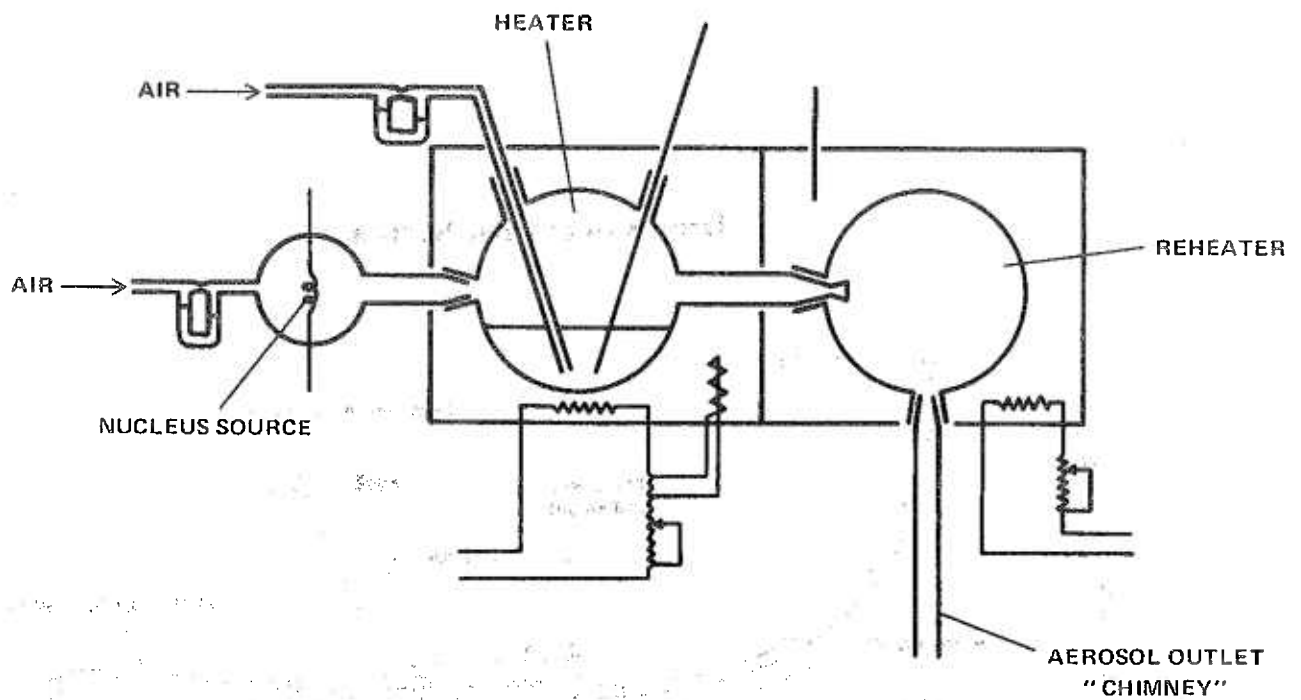


Figure 13. Apparatus for the Production of Large Liquid Drops from a La Mer Generator, Using an Inverted Aerosol Outlet.

Drop Generating System

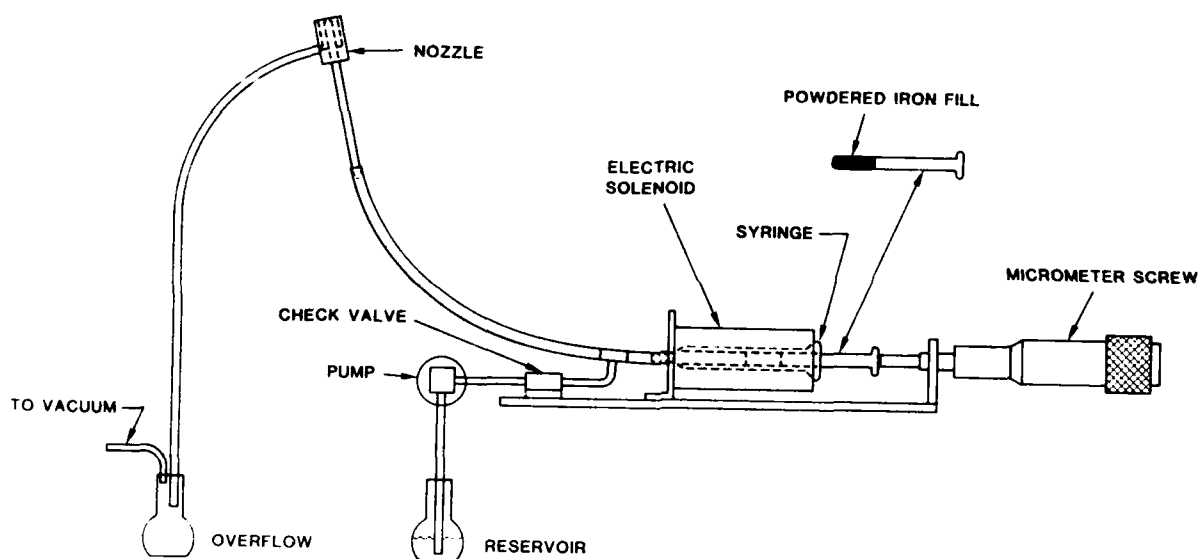


Figure 14. Impulse Drop Ejecting Device of Hoffer and Pitter (1984), as Modified in 1987.

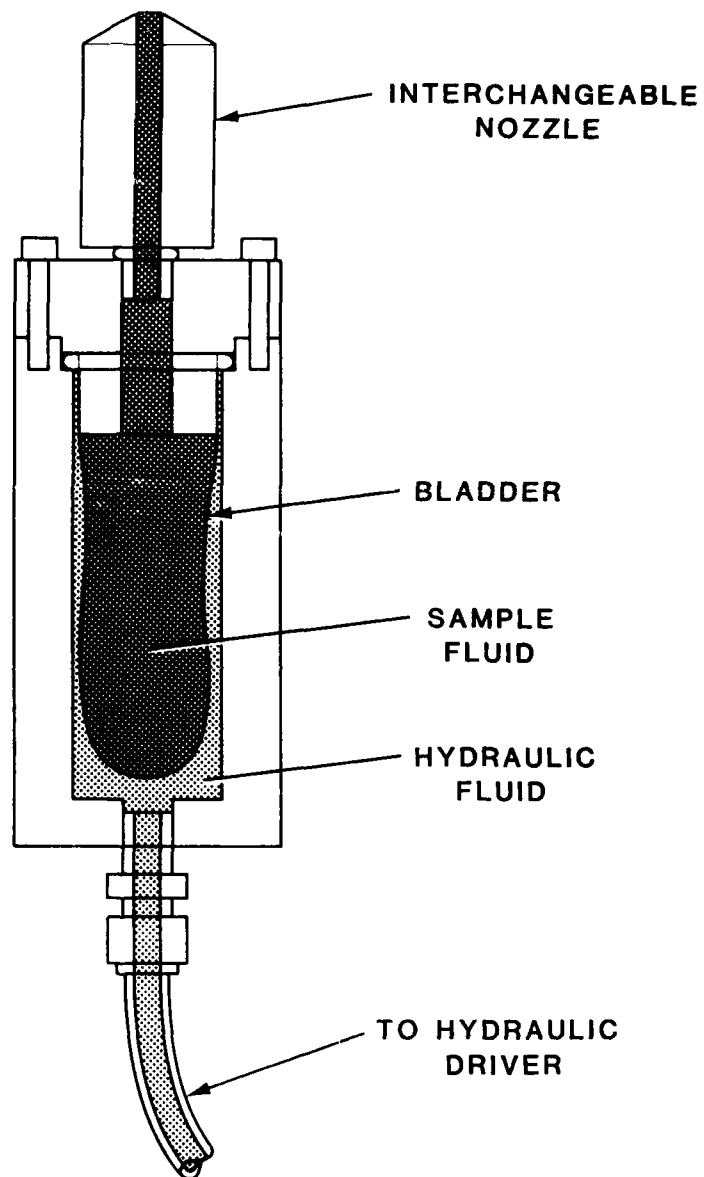


Figure 15. Detail of Two-Fluid Interface of Hoffer and Pitter Impulse Drop Generator, as Modified in 1987.



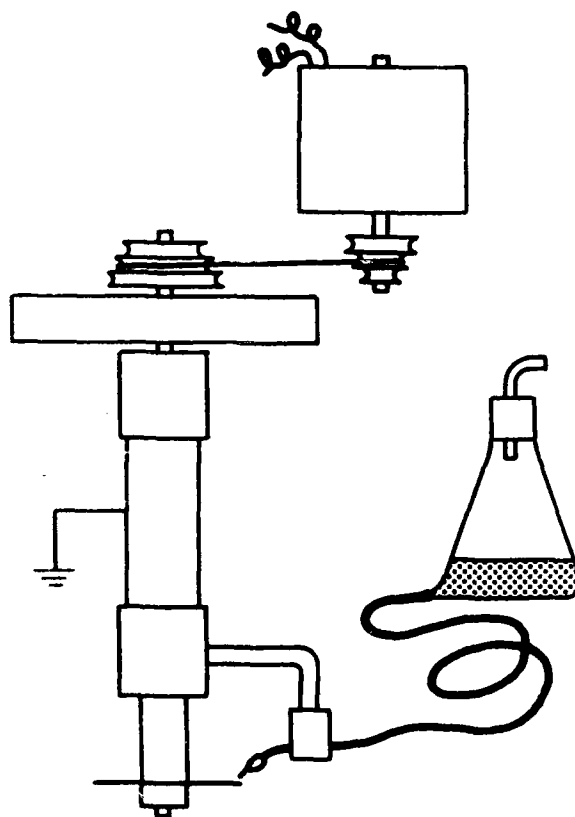


Figure 17. Rotary Blade Drop Separating Device of Rayner and Haliburton (1955).

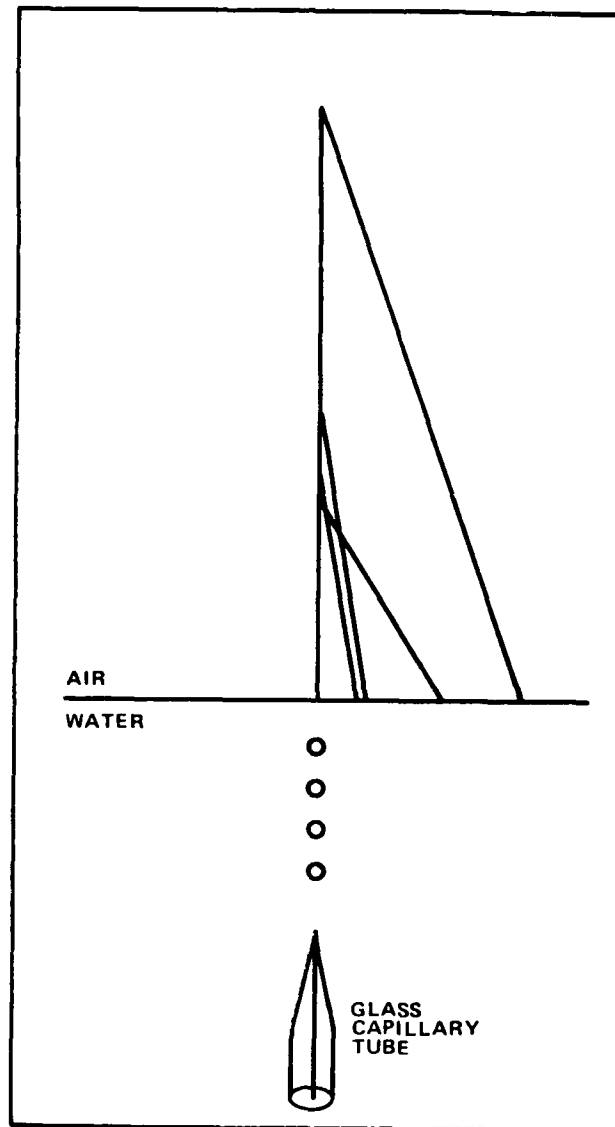


Figure 18. Diagram of Blanchard (1954) Bursting Bubble Technique for Drop Generation, With Lines Representing Various Settling Trajectories of Drops of Various Sizes in a Uniform Airflow from Left to Right.

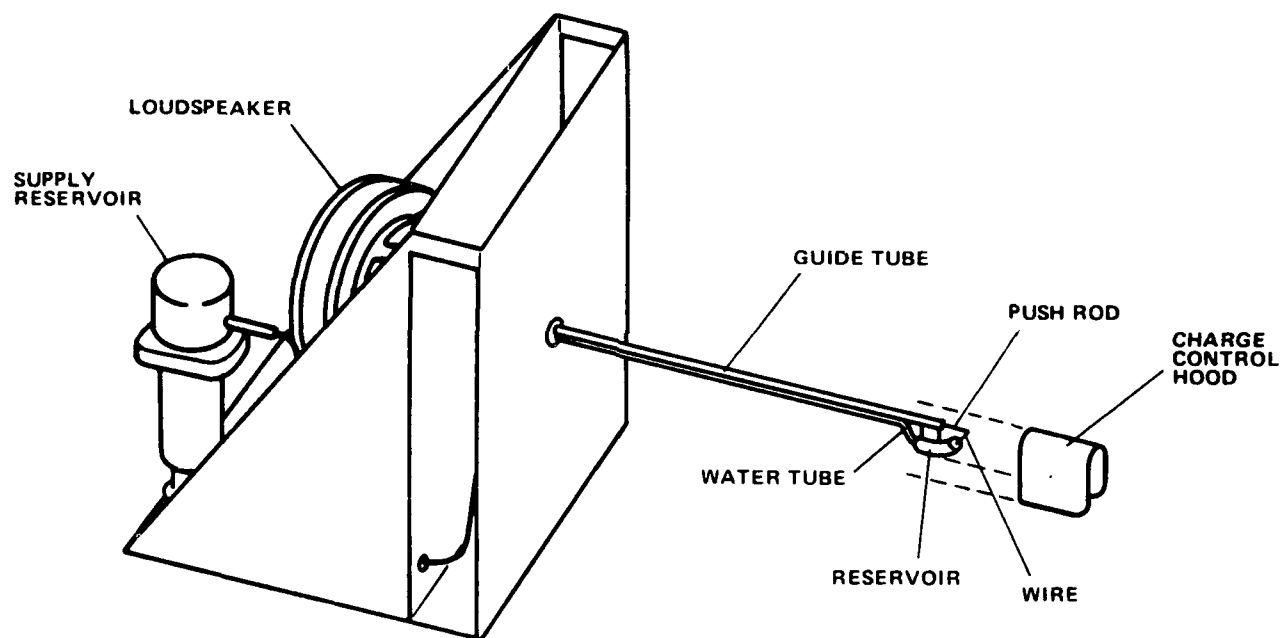


Figure 19. Drop Generator of Abbott and Cannon (1972), Which Pulls a Filament of Liquid from a Drop Surface.

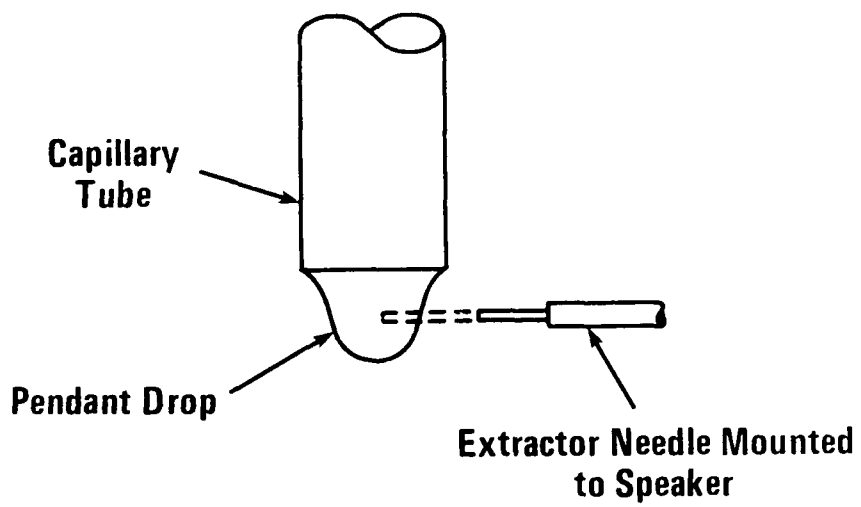


Figure 20. Diagram of National Aeronautical Establishment Drop Generator.

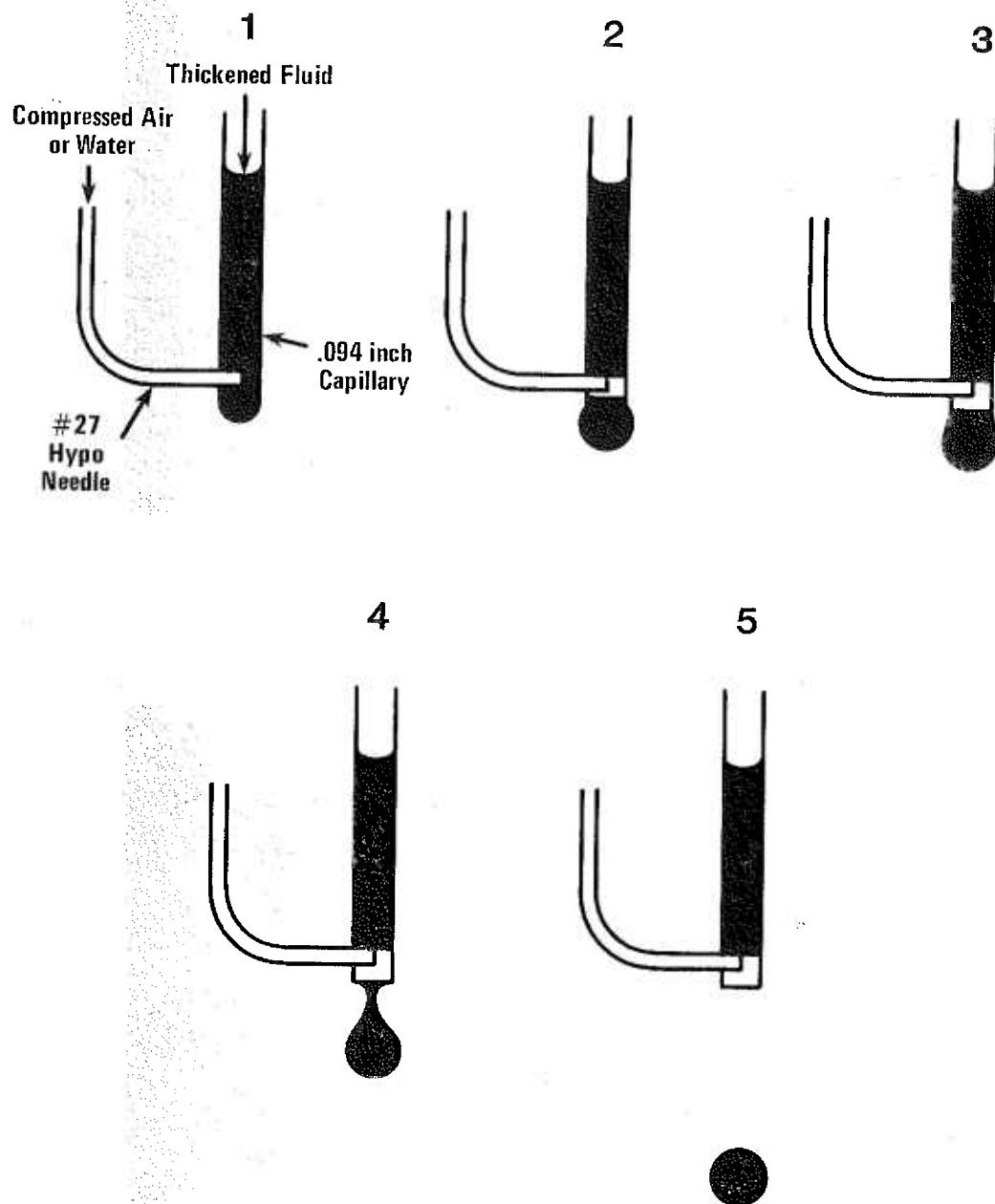


Figure 21. Diagram of DICE, Which Uses the Air Bubble Method of Separating a Visco-Elastic Drop, from Saylor (1985).

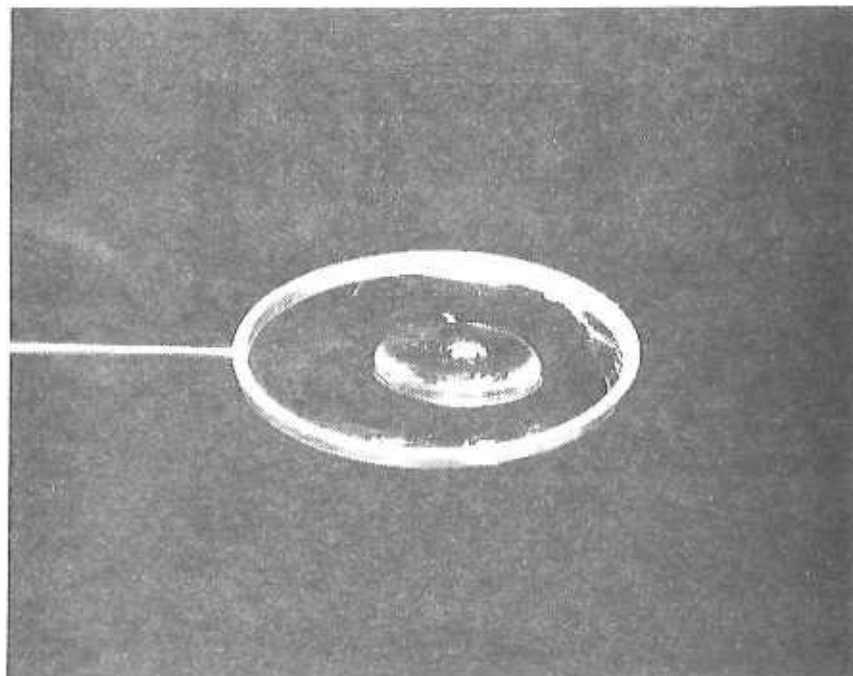
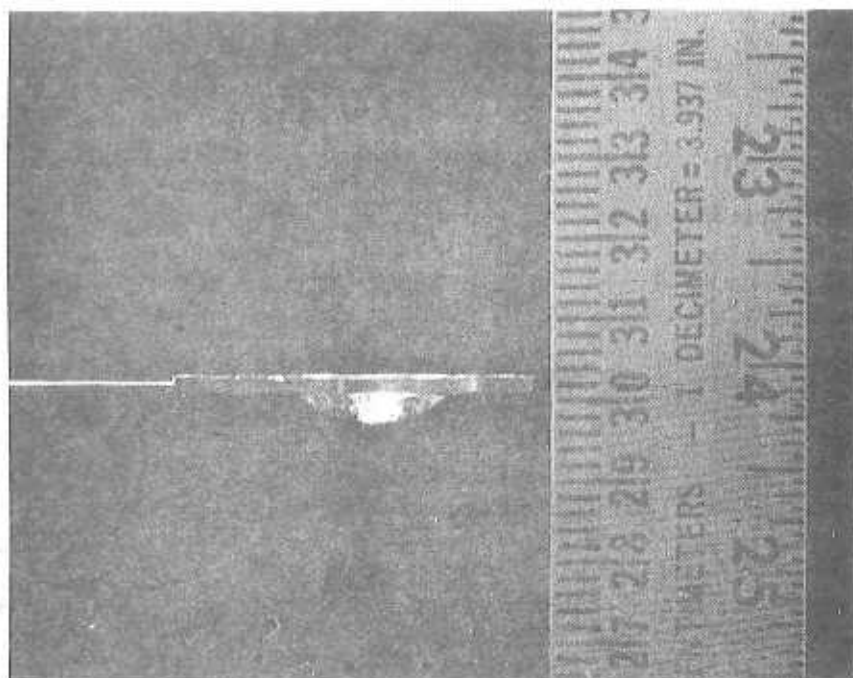


Figure 22. Two Views of the Microfilm Drop Generator of Pitter and Hoffer (1987).